

Remarks:

Claim 1 has been amended. New claim 21 has been added. Claims 2 and 6 were cancelled in a previous Response(s). Claims 12-20 have been withdrawn. Claims 1, 3-5, 7-11, and 21 are presented for the Examiner's review and consideration. It is noted that all nanotubes mentioned herein are carbon nanotubes, unless otherwise noted. Applicants believe the claim amendments and the accompanying remarks presented herein serve to clarify the present invention and are independent of patentability. No new matter has been added.

Amendment to the Specification

No new matter has been added by the amendment to the specification made herein. The specification has been amended only for correction of inadvertent typographical errors.

Amendments to the Claims

No new matter has been added by the amendments to claim 1 made herein. Claim 1 has been amended to clarify that the polymeric material is charged. This amendment is supported by the specification as originally filed, for example, at paragraph [0024] of the published application (paragraph [0022] as filed). Claim 1 has also been amended to clarify that no surfactants, other than the charged polymeric material, are added to the suspension. This amendment is supported by the specification as originally filed, for example, at paragraph [0027] (paragraph [0025] as filed) and Figure 3 of the published application. Furthermore, paragraph [0022] (paragraph [0020] as filed) of the published application, indicates that a water-soluble polymer and a powder of as-produced nanotubes, containing bundled arrays of ropes, are mixed in water or an aqueous solution. Claim 1 has been further amended to clarify that a ready-for-use suspension of carbon nanotubes is obtained by carrying out the claimed method. This amendment is supported by the specification as originally filed, for example, at paragraphs

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[0012] and [0022] (paragraphs [0010] and [0012] as filed) of the published application.

No new matter has been added by the addition of new claim 21. This claim has been added to clarify that when carrying out the described method the carbon nanotubes added to the water solution of polymeric material are as-produced and without further chemical modification, such as oxidation and/or filling the nanotubes with metals. This new claim is supported by the specification as originally filed, for example, at paragraph [0022] (paragraph [0012] as filed) of the published application.

Interview with the Examiner

Applicants thank the Examiner for granting an interview to discuss the claimed invention with regard to the outstanding rejection under 35 U.S.C. § 103(a) and for courtesies extended to Applicants' representatives during this telephone interview on Thursday, June 5, 2008.

Information Disclosure Statement

The following journal article was cited incorrectly at page 2 of the information disclosure statement filed on November 12, 2003: S.J. Tans et al. "Room-temperature transistor based on a single carbon nanotube", Nature 386, May 7, 1998, pages 49-52. This article was also referenced incorrectly in the specification at paragraph [0006] of the published application (paragraph [0004] as filed): S.J. Tans et al, Nature 386 (1997) 474. The specification has been amended herein to correct the citation as follows: S.J. Tans et al. "Room-temperature transistor based on a single carbon nanotube", Nature 393 (1998) 49.

Applicants respectfully request confirmation from the Examiner that the correct article was considered.

Rejection under 35 USC § 103(a)

Claims 1, 3-5, and 7-11 were rejected by the Examiner under 35 USC § 103(a) as allegedly being unpatentable over U.S. Patent 5,114,477 to Mort et al. (hereinafter "Mort") in view of Satishkumar et al. (J. Phys. B: At. Mol. Opt. Phys. 29: 4925-4934 1996; hereinafter "Satishkumar"), Ausman et al. (The Journal of Physical Chemistry 104(38): 8911-8915 2000; hereinafter

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“Ausman”), Qian et al. (Applied Physics Letters 76(20): 2868-2870 2000; hereinafter “Qian”), Sinha et al. (Chemical Physics Letters 332:455-460 2000; hereinafter “Sinha”), and Liu et al. (Science 280:1253-1256 1998; hereinafter “Liu”). For both reasons set forth below and reasons provided in prior Responses, Applicants respectfully disagree with all of the assertions and conclusions of the Examiner and submit that this rejection should be withdrawn.

It is noted that the references are described separately only to clarify what each reference teaches and not to argue each reference separately.

The teachings of Mort, Satishkumar, and Ausman are applied as in the previous Response, filed on April 2, 2008.

Qian

Qian discloses solid, polymer-nanotube composites, in particular, a composite of polystyrene and multiwall carbon nanotubes (MWNT-PS composites). The nanotubes are dispersed homogeneously throughout the polystyrene matrix to form films. These composite films are made by employing a simple solution-evaporation method (using toluene) assisted by high-energy sonication. Qian defines optimum sonication time as the shortest time to disperse the nanotubes into the polystyrene, as longer sonication times has the potential to introduce defects into the nanotubes. Qian aims to understand the tensile fracture mechanisms of the MWNT-PS composites by carrying deformation studies (initiation of cracks in the material) with transmission electron microscopy. *See* abstract and pages 2868-2869. The addition of even a small amount of nanotubes to the polystyrene significantly increases mechanical properties of the polymer. *See* page 2870. Qian does not teach single-walled nanotubes.

Sinha

Sinha teaches a method for preparation of multiwalled carbon nanotubes (MWNTs) filled with metal, in particular cobalt. In this catalytic chemical vapor deposition (CCVD) method, Sinha teaches replacement of the conventional zeolite Na-Y supports with aluminophosphate. The aluminophosphate framework is electroneutral while the zeolite Na-Y framework is highly charged,

so the nature of the metal-support interaction has a strong bearing on the properties of the nanotubes produced. MWNTs are formed in high yields and are easily separated from aluminophosphate-based supports with mild acid treatment and sonication. *See abstract and introduction section (1).* The MWNTs are synthesized by catalytic decomposition of acetylene in a temperature range of 873-1073 K over supported cobalt catalysts. *See experimental section (2).* Weak metal-support interaction in the case of cobalt metal supported on aluminophosphate results in nanotubes filled with metal nanoparticles and nanowires while strong metal-support interaction in the case of cobalt metal supported by zeolite Na-Y results in nanotubes completely free of metal. *See conclusion section (4).* These metal-filled MWNTs have magnetic properties and can be used in magnetic fluids and inks. *See introduction (1) and conclusion (4) sections.* Sinha does not discuss metal-filled or unfilled single-walled nanotubes.

Liu

Liu teaches a method for cutting aggregated ropes of single-walled nanotubes (SWNTs) into short, open-ended pieces called “fullerene pipes” in 100-300 nanometer lengths. *See abstract.* The starting material (raw, single-walled fullerene nanotube felt) was first purified. See page 1253, right column and Figure 1A. The purified material is sonicated in a mix of concentrated acid (sulfuric and nitric), etched in the acid mix, and incubated at 70°C. Liu also teaches a mixture of sulfuric acid and hydrogen peroxide. *See page 1254, all columns.* Liu indicates that the sonication period lasts for days. *See page 1253, right column.* The fullerene pipes produced by this method flocculate (precipitate) rapidly in aqueous solution. Stable, colloidal suspensions in water can be made only with the assistance of surfactants. *See paragraph bridging pages 1254-1255.* Furthermore, the fullerene pipes are shown to remain in van der Waals contact and are aggregated when electrodeposited from the suspension to HOPG. *See Figure 2 and page 1255, center column.* Liu also teach derivatization of the fullerene pipes, for example tethering gold particles to the open ends. *See abstract, page 1255, and Figure 5.*

Argument

First, Applicants note that the current Office Action has been presented in two parts; a Response to Arguments and a rejection of claims 1, 3-5, and 7-11 under 35 USC § 103(a) that is essentially the same as that presented in the final Office Action of January 7, 2008 with the addition of three new references.

At pages 4 and 5 of the “Response to Arguments”, the Examiner again makes reference to the passage from the Bandyopadhyaya article, which states: “The dispersion method is based on an ancient Egyptian recipe, first used 5000 years ago for preparation of carbon-black ink.¹⁹” Footnote 19 references an Encyclopedia Britannica from 1966. This article mentions early inks made from grinding lampblack and a solution of glue or gums, and dispersing this mixture in aqueous solutions to make ink. Based upon this article, the Examiner concludes that Applicants (and the Egyptians) were clearly aware of ink making procedures which dispersed carbonaceous particles in aqueous solution, and furthermore Applicants were aware that dispersions employing Gum Arabic have been known for some time.

The cited article refers to lampblack and not to nanotubes or fullerenes. One of skill in the art knows that the behavior of a particle in aqueous solution depends upon the size, shape, and surface structure of the particle. Lampblack is an amorphous carbon. Even based upon the name alone, amorphous carbon is known to be without form, and clearly lacking the distinct geometric structure of a nanotube or even a fullerene. Thus, one would not draw an immediate parallel between the behavior of lampblack and the behavior of nanotubes in solution.

Additionally, the Examiner himself notes that it is the claims that define the invention (page 3 of the current Office Action). Applicants agree and point out that the claimed method is not limited to making ink, and furthermore, the pending claims do not recite a method for making ink or even mention ink at all. Furthermore, the invention as currently claimed, is not applicable as a method for making ink. This point will be further addressed below.

At page 3 of the “Response to Arguments”, the Examiner asserts that Applicants have not addressed the motivation articulated in the final Office Action of January 7, 2008 and have traversed

the rejection on the basis of differences between fullerenes and nanotubes when the rejection was tailored to the similarities between fullerenes and nanotubes.

First, Applicants respectfully submit that it is perfectly acceptable to address differences, as any differences between the claimed invention and the prior art may be expected to result in some differences in properties between fullerenes and nanotubes. Differences in properties are very useful for distinguishing an invention from prior art and for establishing lack of motivation.

Next, Applicants note that the same alleged “motivation” presented in the final Office Action mailed on January 7, 2008, is repeated in the current Office Action and will be addressed below.

The Examiner again applies Mort as the main reference to support the supposed “obviousness” of the instant application. Additionally, the Examiner states yet again that Mort is the first patent to issue employing carbon nanotechnology. Whether Mort or another was the first patent to issue employing carbon nanotechnology is of no consequence in the current case. However, Applicants point out that methods for destroying ballistic missiles using the sharp edges of fullerenes are taught by U.S. Patent 4,922,827, issued on May 8, 1990.

The Examiner asserts that Mort still forms the basis of a proper rejection, as it discloses the aqueous dispersions as claimed, sans nanotubes. Applicants note that without nanotubes, it is not the dispersion as claimed.

The crux of the Examiner’s argument is still the alleged similar chemistry that can be performed with fullerenes and nanotubes. Thus, the Examiner continues to maintain that fullerenes and carbon nanotubes can be used interchangeably for the same purposes and results and that one can simply replace Mort’s fullerenes with nanotubes and arrive at the claimed invention. Applicants respectfully disagree with the Examiner’s assertions and conclusions.

Applicants do not deny that fullerenes and nanotubes have many features in common, however, it is their differences in properties which render the claimed method unobvious in light of the prior art.

These two carbon forms are related in that nanotubes are derived from fullerenes. For example, a single- wall C₆₀ –derived tube can be formed by bisecting a C₆₀ molecule at the equator and joining the two resulting hemispheres with a cylindrical graphite tube. *See* page 757 of Dresselhaus et al. (cited on the information disclosure statement filed on November 12, 2003). However, the nanotube has a different shape and structural geometry from that of a fullerene. The nanometer diameter and millimeter length of the tube confers distinct properties to the nanotube. For example, with regard to elasticity, the strain energy per carbon atom for fullerenes of similar diameter is much larger than for the comparable nanotubes. This larger strain energy for the fullerenes reflects their two-dimensional curvature in comparison to tubules which have a one-dimensional curvature. *See* page 857 of Dresselhaus et al.

Accordingly, the fullerenes of Mort and the carbon nanotubes of the claimed method are structurally and chemically different materials; *i.e.* they differ in the number and arrangement of atoms and thus, have differing physico-chemical properties. These materials are prepared by different methods, and are intended for different uses. Mort, following the tradition of using carbon derivatives (graphite, soot) for writing and printing, broadens the use of carbon derivatives to include fullerenes and provides an ink including fullerenes as a colorant. The scope of the carbon structures to be used as colorants is defined by Mort as spherical shells of about 32 to 1,000 carbon atoms, prototypically a fullerene having 60 atoms arranged in a truncated icosahedron. *See* column 3, lines 37-45. As they are black in color, and further being soluble in organic solvents, the fullerenes are proposed to be used in inks for thermal ink jet applications. *See* column 4, lines 17-19. These fullerenes provide inks by simple mixing with solvents. *See* column 8, lines 40-56. Both experimental examples disclosed by Mort exemplify the preparation of homogeneous solutions. *See* column 8, lines 45-51 and column 9, lines 19-68.

The Examiner notes that Mort discloses the ability to disperse pigments as large as 0.1 micron (100 nanometers) and thus, Mort would be perfectly capable of dispersing the nanotubes of the size described by Applicants. The Examiner further notes that Liu teaches nanotubes in the range of 100-300 nanometers. From these observations, the Examiner concludes that “size has no

relevance to the claims” and further, “there is no reason to believe that longer nanotubes would behave differently.” *See* page 8 of the current Office Action.

Applicants respectfully disagree. The issue is whether a spherical particle, as disclosed by Mort, and a nanotube, as disclosed by Applicants, can be expected to exhibit similar properties in solution, as the Examiner appears to believe. Mort discloses particles having a diameter of 100 nanometers. A nanotube may also have a diameter of 100 nanometers. However, as noted above, a nanotube also has a length in millimeters.

Carbon nanotubes are macromolecular threads with a typical diameter of a few nanometers and a length of up to 10 microns. *See* paragraph [0005] of the published application. In contrast fullerenes would have a length of about 0.001-0.003 microns. The fibers of separate carbon nanotubes have a thickness of 2-3 nanometers, corresponding to 20-30 \AA , while the fullerene spheres have a diameter of 10 \AA . *See* paragraph [0035] and Figure 4 of the published application. Carbon nanotubes are entangled in ropes and networks due to strong van-der-Waals interactions which may strongly bind 100-500 single tubes, up to 1500 \AA . *See* paragraph [0009] of the published application. Due to such aggregation, carbon nanotubes are insoluble in both water and organic solvents, and are even difficult to disperse in solvents. *See* Ausman.

Furthermore, considering rheology and contact forces, one of skill in the art knows that the behavior of a particle in aqueous solution depends upon the size, shape, and surface structure of the particle. There is not much in common between the behavior in liquids of small spherical molecules of distinct geometry and tube-like, nearly macroscopic fibers. Thus, one of ordinary skill in the art would not draw any parallel between the behavior of fullerenes and the behavior of nanotubes in solution.

Further, with regard to solubility, nanotubes would not be expected to form a real solution, but rather, to form unstable suspensions of aggregated tubes that can block the needles of inkjet printers. Thus, as mentioned above, nanotubes would not be obviously applicable for preparing homogeneous inks like that of Mort. This is further evident in that Mort recognizes that particle size

is important to prevent clogging of the ink channels in printers. *See* column 7, lines 46-50. Even if one of ordinary skill in the art were to replace the fullerenes of Mort with the nanotubes of the invention, the resulting product would have different properties from that of Mort's original solution. Thus, fullerenes and nanotubes are not exchangeable for the same results.

Fullerenes and nanotubes, described respectively in Mort and in the claimed method, differ in ability to form suspensions and solutions. Further, Mort introduces fullerenes into liquid in an entirely different manner from that of the claimed method. Mort prepares the ink solution by simple mixing of the ingredients and by discarding insoluble material (filtering). *See* Mort column 8, lines 42-45. In contrast, the instant inventors prepare the suspension by sonicating the ingredients and discarding the soluble material. *See* paragraph [0023] of the published application. In Mort's method, the ingredients include organic solvent and surfactant and, in contrast, the ingredients in the claimed method include water with a charged polymer. *See* Example 1 of Mort and paragraph [0026] of the published application. Considering all of these differences, it is unlikely that Mort would lead one of ordinary skill in the art to the claimed method.

The Examiner asserts that the addition of a surfactant has no relevance to patentability. *See* page 2 of the current Office Action. Applicants respectfully disagree. Claim 1 has been amended herein to clarify that no surfactants, other than the charged polymeric material, are added to the suspension. As noted above, Ausman discloses the insolubility of carbon nanotubes in water and organic solvents. Liu discloses that stable, colloidal suspensions of nanotubes in water can be made only with the assistance of surfactants. *See* paragraph bridging pages 1254-1255. Accordingly, the prior art teaches insolubility of carbon nanotubes in water without the aid of surfactants. The claimed method provides a suspension of separated, carbon nanotubes in water without the use of surfactants or chemical modifications. Since the claimed method accomplishes a solubility thought to be unachievable in the prior art, the lack of surfactants has relevance to patentability.

The Examiner asserts that sonication is common knowledge, and notes the generic mixing step of Mort. *See* pages 3 and 7 of the current Office Action. Applicants assert that a "simple or generic mixing step" would not necessarily lead one of ordinary skill in the art to sonication. For

example, sonication can be more intense and may incorporate too much energy into a system and negatively affect its properties. *See*, for example, Liu at page 1254. Nevertheless, Applicants do not state either that sonication is a novel method, or that sonication is a novel step in mixing carbon nanotubes. However, in the method as currently claimed, sonication combined with other known techniques, provides a ready-for-use dispersion of separated carbon nanotubes, in a concentration and stability that have never been achieved before.

Applicants respectfully submit that the statements the Examiner refers to as “similarity” or “motivation” are irrelevant to the invention as claimed. For example, at page 4 of the final Office Action mailed on January 7, 2008, and page 7 of the current Office Action, the Examiner states that Mort identifies the numerous advantages of using fullerenes, including their solubilities in organic and aqueous solvents (column 4, lines 17-23). And further the ability to tailor the properties of the fullerene through chemical modification was cited as advantageous (column 4, lines 25-34). Additionally, in the current Office Action at page 7, the Examiner cites Satishkumar (pages 4927 and 4930), and asserts that the same properties that make spherical fullerenes advantageous are well described in the literature. And even further, the Examiner asserts that Sinha provides explicit motivation by noting that nanotubes are useful as magnetic ink.

The first passage of Mort, cited by the Examiner, actually refers to advantages of fullerenes specifically for liquid thermal ink jet applications, including solubility in organic solvents and water. However, to obtain solubility in water, chemical modification is required by attachment of diamine chromophores to the surface of the carbon shell. *See* column 4, lines 17-23. Further, the tailored properties are dependent on chemical modification of the carbon shell. *See* column 4, lines 25-34. Additionally, Mort specifically notes that other forms of carbon (diamond and graphite) are not considered to be soluble in these solvents. *See* column 4, lines 23-25. This statement indicates that different forms of carbon were not expected to exhibit the same solubilities.

The Examiner further asserts, at page 4 of the final Office Action mailed on January 7, 2008 and page 7 of the current Office Action, that the same properties that make fullerenes advantageous

are well described in the literature for nanotubes (Satishkumar, filling 4927 and functionalizing 4930).

Satishkumar discloses two methods directed to nanotubes; opening, filling with metals, and closing the tubes and functionalizing the tubes by treatment with acid. Sinha also discloses methods for filling nanotubes with metals. Mort does not discuss any of these methods. Thus, the methods and properties of Satishkumar and Sinha cannot be said to be merely the methods of Mort disclosed for nanotubes, as the methods of Satishkumar and Sinha are unrelated to those of Mort. There is nothing in Satishkumar, Sinha, or any other combination of prior art that would motivate an artisan of ordinary skill to practice the method of Mort with nanotubes, especially when Mort acknowledges that all forms of carbon are not expected to have the same solubility. *See* column 4, lines 23-25. Regardless, the instant invention does not rely on organic solvents or chemical modification of nanotubes. The starting material of the claimed method is unmodified, as-prepared nanotubes.

A rationale to support a conclusion that a claim would have been obvious is that all the claimed elements were known in the prior art and one skilled in the art could have combined the elements as claimed by known methods with no change in their respective functions, and the combination would have yielded nothing more than predictable results to one of ordinary skill in the art. *See* MPEP 2143.02. As discussed above, the substitution of Mort's fullerenes with nanotubes of the instant invention would result in neither the claimed invention nor the original solution. Furthermore, such a substitution would render Mort's solution unusable for its intended purpose as an ink, *i.e.* nanotubes would aggregate and clog the ink jets of the printer. The instant invention demonstrates that highly insoluble carbon nanotubes can form stable, concentrated dispersions in water, in the presence of a hydrophilic polymer, without reliance on surfactants and/or chemical modification. This type of dispersion was not achieved until the instant invention. Thus, the claimed method is neither predictable or obvious.

Applicants respectfully submit that the cited references (Mort, Satishkumar, Ausman, Qian, Sinha, and Liu) cannot be combined to provide the claimed method or products that result from carrying out the claimed method. Mort does not teach carbon nanotubes or solubility of fullerenes

without the help of surfactants, solvents, and/or chemical modification. Satishkumar, Ausman, Qian, Sinha, and Liu do not remedy the deficiencies of Mort. Satishkumpar teach chemical derivatizations and metal-filled nanotubes and do not lead in the direction of charged polysaccharides. Sinha also teach metal-filled nanotubes. Ausman does not teach how to perform solution-based handling of nanotubes without the use of organic solvents. Qian teach solid, polystyrene-nanotube composite films. Liu teach a method for cutting longer nanotubes into shorter pipes and disclose that the shorter pipes precipitate in water without the addition of surfactants. As established above, the claimed method can not be derived from simply replacing Mort's fullerenes with carbon nanotubes. At the time of the invention nanotubes were not expected to be soluble without the aid of surfactants, solvents, and/or chemical modification. None of the cited references teach the solubility of carbon nanotubes in water with a hydrophilic polymer. A person of ordinary skill in the art, having the cited references in front of him/her, would not be inspired or motivated to add carbon nanotubes to a water solution of hydrophilic polymer and expect to produce a stable, concentrated dispersion of single tubes without reliance on surfactants and/or chemical modification, because the combination of teachings of the cited references and/or other knowledge in the art indicate that it cannot be done. Many prior artisans have attempted to comply with the hydrophobic character of the carbon nanotubes by introducing organic solvents or surfactants, or at least by increasing the hydrophobicity of the dispersing agents, but no one has tried the apparently futile procedure according to the instant invention. The instant inventors were the first to provide a simple method (short mixing periods and limited use of surfactants) for unbundling carbon nanotubes in aqueous solutions of hydrophilic polymers resulting in a stable dispersion (of carbon nanotubes) that can be dried into a ready-for-use powder of individual carbon nanotubes. The unbundling and stabilization of the single tubes is demonstrated unequivocally in the specification by a combination of X-ray scattering and cryo-TEM imaging. No such method or result is taught or suggested by the combination of the cited references (Mort, Satishkumar, Ausman, Qian, Sinha, and Liu) or any other prior art for carbon nanotubes or fullerenes.

Accordingly, independent claim 1 would not be obvious to one of ordinary skill in the art reading Mort in view of Satishkumar, Ausman, Qian, Sinha, and Liu under 35 U.S.C § 103(a). As

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claims 3-5, 7-11, and 21 depend from claim 1, these dependent claims necessarily include all of the elements of the base claim. Thus, Applicants respectfully submit that the dependent claims are allowable over the cited references for at least the same reasons.

In light of all of the foregoing, Applicants request reconsideration and withdrawal of the rejection of claims 1, 3-5, and 7-11 under 35 U.S.C § 103(a).

Conclusion

In light of the foregoing amendments and remarks, this application is now in condition for allowance and early passage of this case to issue is respectfully requested. If any questions remain regarding this amendment or the application in general, a telephone call to the undersigned would be appreciated since this should expedite the prosecution of the application for all concerned. The fee for submission of an information disclosure statement pursuant to Section 1.17(p) in the amount of \$180 is believed to be due and is being paid by credit card. No additional fee is believed due at this time. However, please charge any other required fee (or credit any overpayments of fees) to the Deposit Account of the undersigned, Account No. 500601 (Docket No. 7640-X03-011).

Respectfully submitted,

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